CONTINUED STUDY OF PRESOLAR GRAPHITE FROM MURCHISON SEPARATE KFA1. S. Amari<sup>1</sup>, F. J. Stadermann<sup>1</sup>, E. Zinner<sup>1</sup> and R. S. Lewis<sup>2</sup>, <sup>1</sup>Laboratory for Space Sciences and the Physics Department, Washington University, One Brookings Dr., St. Louis, MO 63130, USA (sa@wuphys.wustl.edu, fjs@wuphys.wustl.edu, ekz@wuphys.wustl.edu), <sup>2</sup>Enrico Fermi Institute, University of Chicago, 5630 S. Ellis Ave., Chicago, IL 60637, USA (r-lewis@uchicago.edu)

**Introduction:** Presolar graphite was isolated from the Murchison meteorite [1] only 3 years after the discovery of presolar SiC [2]. Yet it has not been studied as extensively as SiC. One of the reasons is that trace element concentrations in graphite grains are, in general, not high enough to perform isotopic analysis with sufficient precision. A new type of ion microprobe, the NanoSIMS, has recently been installed at Washington University in St. Louis. Its high sensitivity at highmass resolution and its multi-detection capability [3] make it possible to measure isotopic ratios of trace elements with unprecedented precision. We have previously reported C, N, O, and Si isotopic ratios of graphite from the Murchison KFA1 separate (2.05-2.10g/cm<sup>3</sup>) [4]. We extended our studies to Mg-Al, K, Ca, and Ti in the same KFA1 graphite grains that we had previously analyzed in order to obtain correlated isotopic ratios.

**Experimental Procedures:** The measurements were made with the NanoSIMS at Washington University. We analyzed Mg-Al ratios in 75 grains, K-Ca isotopes in 24 grains, and Ti isotopes in 32 grains. An O primary beam was rastered over a 3×3 µm² area on the grains. For Mg-Al analysis, 12°C, 24°M g, 25°M g, 26°Mg and 27°Al were simultaneously collected with five small electron multipliers. Potassium and Ca isotopic analysis was performed in the combined analysis mode, which uses magnetic peak jumping in connection with multi-detection. In the first step, 12°C, 39°K, 41°Ca, 42°Ca, 44°Ca, and 48°Ti after changing the magnetic field and deflection voltages of the electron multipliers. Titanium isotopes were also analyzed in the combined mode: 12°C 46°Ti, 48°Ti, 50°Ti, and 52°Cr in the first step and 47°Ti, 49°Ti, 51°V, and 53°Cr in the second step. Murchison matrix was used as standard for Mg and K, and a mixture of perovskite and chromite was used for Ca, Ti and Cr as well as for centering 51°V.

**Results and Discussion:** *Mg-Al.*  $^{26}$ Al/ $^{27}$ Al ratios were calculated from  $^{26}$ Mg excesses, where  $\delta^{25}$ Mg/ $^{24}$ Mg values were subtracted from  $\delta^{26}$ Mg/ $^{24}$ Mg values under the assumption that nuclear processes (such as neutron capture) would produce approximately equal excesses in the neutron-rich Mg isotopes [5-8]. Of 22 grains with detectable  $^{26}$ Al, 14 grains have inferred  $^{26}$ Al/ $^{27}$ Al ratios higher than  $1\times10^{-2}$ , 11 of those have  $^{12}$ C/ $^{13}$ C ratios lower than solar (Fig. 1). Interestingly, also higher  $\delta^{25}$ Mg/ $^{24}$ Mg values are observed in grains with  $^{12}$ C/ $^{13}$ C ratios lower than solar (Fig. 2). This is puzzling because grains with high  $^{12}$ C/ $^{13}$ C ratios, which are likely to be of a supernova origin, are expected to have high  $\delta^{25}$ Mg/ $^{24}$ Mg values: in the He/N zone of supernovae [5, 6],  $^{25}$ Mg is destroyed via the  $^{25}$ Mg(p,γ)  $^{26}$ Al reaction and the  $^{12}$ C/ $^{13}$ C ratio is expected to be low (8 for 15M $_{\odot}$  supernovae) due to the CNO cycle, while in the He/C zone as well as in the O/C and

O/Ne zones  $^{25}Mg$  is produced by neutron capture. The  $^{12}C/^{13}C$  ratio in the He/C zone is extremely high (10<sup>5</sup>) due to the triple-areaction. Thus, to obtain lower  $^{12}C/^{13}C$  ratios, material from the He/N zone had to be incorporated into the mix from which the grains formed (if they indeed formed in supernovae), bringing down  $\delta^{25}Mg/^{24}Mg$  values. On the other hand, in AGB (asymptotic giant branch) stars,  $^{25}Mg$  and  $^{12}C$  that are produced in the He-burning zone are brought into the envelope during third dredge-up episodes [7, 8]. Thus, in both types of stellar sources  $^{25}Mg/^{24}Mg$  and  $^{12}C/^{13}C$  ratios are expected to be positively correlated.

Ca. Unfortunately, for unknown reasons the analyzed mount was heavily contaminated and isotopically anomalous Ca in some of the grains was undoubtedly swamped by normal Ca. Nonetheless, 14 grains turned out to be anomalous. Seven grains have excesses in <sup>42</sup>Ca, <sup>43</sup>Ca, and <sup>44</sup>Ca, 2 grains in <sup>42</sup>Ca and <sup>44</sup>Ca. The highest 8<sup>43</sup>Ca/<sup>42</sup>Ca value is 691±50%. Three grains have deficits in <sup>42</sup>Ca and one of the three in <sup>43</sup>Ca as well. Eight grains show evidence for the initial presence of short-lived <sup>41</sup>Ca from <sup>41</sup>K excesses; inferred <sup>41</sup>Ca/<sup>40</sup>Ca ratios range from 1×10<sup>-3</sup> to 8×10<sup>-3</sup>. Because of the high level of the Ca contamination, these ratios should be regarded as lower limits.

*Ti.* Ti analysis was also hindered by Ca and Cr contamination. Many grains have lower than solar apparent <sup>47</sup>Ti/<sup>48</sup>Ti ratios, but this is a result of large <sup>48</sup>Ca interferences with <sup>48</sup>Ti<sup>+</sup>. Since we did not measure <sup>40</sup>Ca during the Ti isotopic analysis, we tried to correct for the <sup>48</sup>Ca interference from <sup>40</sup>Ca counts obtained during the K-Ca analysis and by assuming the <sup>48</sup>Ca/<sup>40</sup>Ca ratio to be solar. We discarded the data if the inferred <sup>48</sup>Ca/<sup>48</sup>Ti ratio was larger than 0.1. Of the 24 grains, only 7 grains had acceptably low <sup>48</sup>Ca/<sup>48</sup>Ti ratios. If no K-Ca analysis was made, grains with large negative δ<sup>47</sup>Ti/<sup>48</sup>Ti values (-240 to -60%) were also discarded. Because of the large <sup>50</sup>Cr interference we could not determine <sup>50</sup>Ti/<sup>48</sup>Ti ratios.

All 7 grains with anomalous Ti have <sup>49</sup>Ti excesses ranging up to 1500‰. One of them shows a V-shape isotopic pattern when normalized to <sup>48</sup>Ti, with about equal amounts of excesses in <sup>46</sup>Ti and <sup>49</sup>Ti (1200 and 1500‰). Another grain has a <sup>47</sup>Ti excess of 180‰.

**Stellar Sources.** Because of the contamination, we are certain that at least Si and Ca isotopic ratios of the KFA1 grains in this mount have to be regarded as upper or lower limits, depending on the anomalies. With this constraint in mind, we consider stellar sources of KFA1 graphite.

Low-density graphite grains from Murchison separate KE3 (1.65-1.72g/cm³) have been most extensively studied among the graphite fractions from this meteorite [9]. Many KE3 grains have <sup>18</sup>O and <sup>28</sup>Si excesses

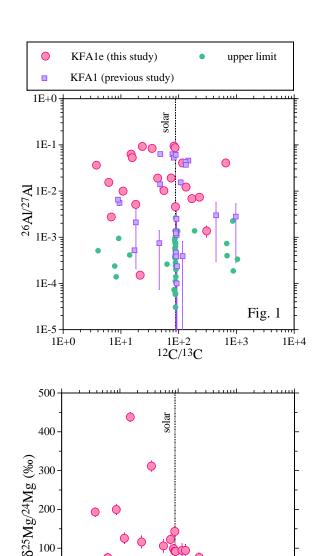
and a few grains show evidence for the initial presence of short-lived <sup>44</sup>Ti (from large <sup>44</sup>Ca excesses). Those isotopic features indicate that they formed in supernova ejecta. Some of the KFA1 grains have <sup>18</sup>O and <sup>28</sup>Si excesses, similar to KE3 grains. A supernova origin can be assigned to these grains.

Of 43 grains analyzed for O isotopic ratios, 22 grains are  $^{16}\mathrm{O}\text{-rich}$ , with  $^{18}\mathrm{O}/^{16}\mathrm{O}$  ratios down to 0.75×solar and  $^{17}\mathrm{O}/^{16}\mathrm{O}$  ratios down to 0.68×solar. In the early Galaxy, isotopes made from α particles such as <sup>16</sup>O (primary isotopes), were enriched relative to those produced from primary isotopes (secondary isotopes). Oxygen in the envelope in low-mass lowmetallicity stars, which were born in the early stages of the Galaxy, are predicted to be <sup>16</sup>O-rich, even after the first dredge-up, and the subsequent evolution of the star does not change the O isotopic composition significantly [10]. <sup>12</sup>C/<sup>13</sup>C ratios of the <sup>16</sup>O-rich grains range from 3.5 to 1400. If these grains were produced in low-mass low-metallicity stars and the stars experienced third dredge-up episodes,  $\delta^{25}$ Mg/ $^{24}$ Mg and  $\delta^{30} \text{Si}/^{28} \text{Si}$  values of the grains should increase along with  $^{12} \text{C}/^{13} \text{C}$  ratios. However, a negative correlation is observed between  $^{12} \text{C}/^{13} \text{C}$  ratios and  $\delta^{25} \text{Mg}/^{24} \text{Mg}$  values (Fig. 2), contrary to what is expected if the grains formed in AGB stars. A better test would be to check whether there exists any correlation between Si and Ti isotopic ratios, because such a correlation has been observed in grains of an AGB star origin [11]. However, Ti data in many grains are of no use due to the overwhelming <sup>48</sup>Ca interference with <sup>48</sup>Ti.

<sup>41</sup>Ca/<sup>40</sup>Ca ratios in the envelope of AGB stars are expected to be on the order of 10<sup>-4</sup> [12] while those in the He/C, O/C and the other O-rich zones of supernovae range up to 0.02 [5, 6]. The observed ratios in the grains are thus consistent with a supernova origin.

In summary, some of the KFA1 grains most likely have formed in supernovae. However, we are not able to infer any other stellar sources, partly because of the high level of contamination of the mount. In the future, we hope to analyze clean mounts to better understand the origin of the KFA1 graphite grains.

References: [1] Amari S. et al. (1990) Nature, 345, 238-240. [2] Bernatowicz T. et al. (1987) Nature, 330, 728-730. [3] Stadermann F. J. et al. (1999) Lunar Planet. Sci., XXX, Abstract #1407. [4] Amari S. et al. (2002) Meteorit. Planet. Sci., 37, A11. [5] Meyer B. S. et al. (1995) Meteoritics, 30, 325-334. [6] Woosley S. E. and Weaver T. A. (1995) Astrophys. J. Suppl., 101, 181-235. [7] Busso M. et al. (1999) Ann. Rev. Astron. Astrophys., 37, 239-309. [8] Karakas A. L. and Lattanzio J. C. (2002) Astrophys. J., submitted. [9] Travaglio C. et al. (1999) Astrophys. J., 510, 325-354. [10] Boothroyd A. I. et al. (1994) Astrophys. J., 430, L77-L80. [11] Hoppe P. et al. (1994) Astrophys. J., 430, 870-890. [12] Wasserburg G. J. et al. (1995) Astrophys. J., 440, L101-L104.



1E+0

1E+1

solar

Fig. 2

1E+4

1E+3

1E+2

12C/13C